



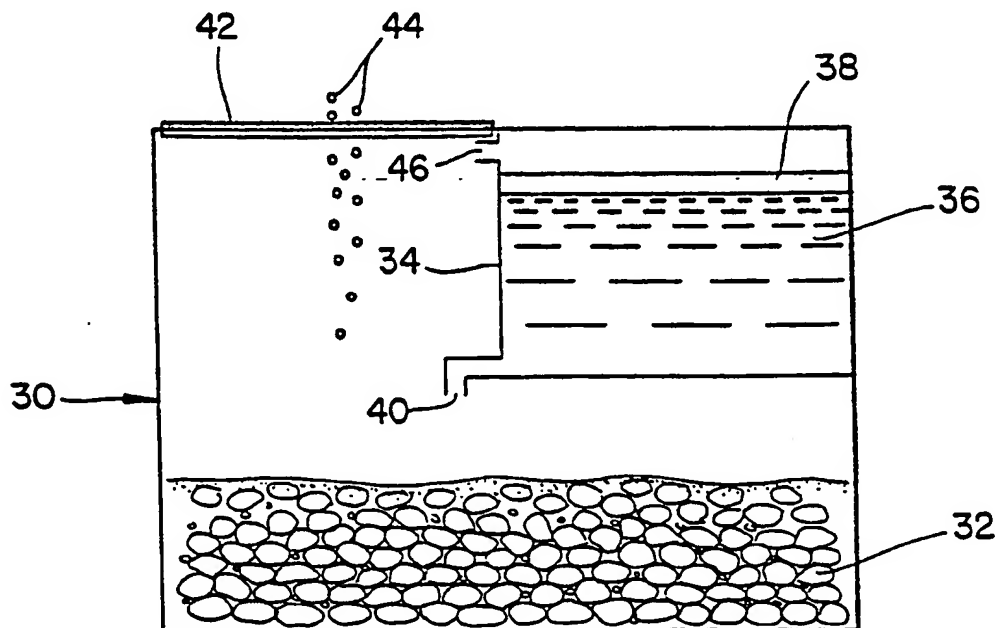
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(21) International Application Number: PCT/US90/01523 (22) International Filing Date: 21 March 1990 (21.03.90) (30) Priority data: 326,791 21 March 1989 (21.03.89) US (71) Applicant: LIM TECHNOLOGY LABORATORIES, INC. [US/US]; 409 East Main Street, Richmond, VA 23219 (US). (72) Inventors: LIM, Franklin ; 6500 Boatwright Drive, Richmond, VA 23226 (US). MOSS, Richard, D. ; 12565 Brook Lane, Chester, VA 23831 (US). (74) Agent: HUNTINGTON, R., Danny; Burns, Doane, Swecker & Mathis, The George Mason Building, Washington & Prince Streets, P.O. Box 1404, Alexandria, VA 22313-1404 (US).		(81) Designated States: AT (European patent), AU, BE (European patent), CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FI, FR (European patent), GB (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), NO, SE (European patent). Published With international search report.

(54) Title: CONTROLLED RELEASE OF DESIRED PRODUCT THROUGH THE CONTROLLED REACTION OF PRECURSOR



(57) Abstract

A method of controlled release of a gas (44) comprising the controlled introduction of a specific reacting or catalyzing solution (36) into contact with a precursor compound (32).

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CONTROLLED RELEASE OF DESIRED PRODUCT
THROUGH THE CONTROLLED REACTION OF PRECURSOR

BACKGROUND OF THE INVENTION

The present invention relates to the controlled release of a desired product, and more specifically, to the controlled release of a desired product through the controlled reaction of a precursor compound.

Oxygen (O_2) for industrial and health care uses comes almost exclusively from liquid oxygen prepared by the distillation of liquid air. In some special instances, limited amounts of oxygen may come from the electrolysis of water. Certain chemicals and chemical reactions can also produce oxygen, although these have never been a practical or regular means of producing oxygen for routine use.

Similarly, carbon dioxide (CO_2) for industrial and health care is recovered from synthetic gas in ammonia production, from substitute natural gas production, from cracking of hydrocarbons, and from natural springs or wells. For laboratory purposes it is obtained by the action of an acid on a carbonate, but this is not a practical way of producing carbon dioxide for routine use.

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Oxygen sources generally involve the use of compressed gas or cryogenic storage tanks or cylinders which are heavy and inconvenient to handle, while electrolysis has the inconvenience of requiring a source of electricity. Carbon dioxide is generally provided in liquid form in steel cylinders which are heavy and inconvenient to handle, or in solid form which requires the use of refrigeration and other special storage conditions. Thus a need exists for a simple convenient source of producing limited amounts of oxygen or carbon dioxide.

SUMMARY OF THE INVENTION

The present invention is a method of controlled release of a gas comprising the controlled introduction of a specific reacting or catalyzing solution into contact with a precursor compound.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a graph illustrating oxygen release from solutions of sodium perborate monohydrate.

Fig. 2 is a graph illustrating oxygen release from solutions of sodium perborate tetrahydrate.

Fig. 3 is a graph comparing oxygen release from solutions of sodium perborate monohydrate when different catalytic solutions are employed.

Fig. 4 is a graph illustrating controlled release of oxygen from sodium perborate.

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Fig. 5 is a schematic illustration of an apparatus for use in practice of the present invention.

Fig. 6 is a schematic illustration of another apparatus for use in the practice of the present invention.

Fig. 7 is a schematic illustration of yet another apparatus for use in the practice of the present invention.

Fig. 8 is a graph illustrating controlled release of oxygen from urea peroxide.

DETAILED DESCRIPTION OF THE INVENTION

The present invention deals with a method of producing controlled release of a desired product or material through the combination of the reaction of a precursor compound with a specific reacting or catalyzing solution and the controlled introduction of the said specific reacting or catalyzing solution into a reaction-producing contact with the precursor compound. This mechanism of producing controlled release can be depicted as follows:

+ specific catalyzing solution or
|
| reacting system at controlled rate
|
|
|
↓
Precursor compound ----> Controlled release
of desired product

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In a preferred embodiment, the present invention takes the form of a unit which contains a peroxygen compound which is a precursor compound of O_2 , or a carbonate or bicarbonate compound, which are precursor compounds of CO_2 . The controlled release of the desired product in this embodiment is O_2 or CO_2 which is control-released by the controlled introduction into contact with the respective precursor compound of an appropriate catalyst or reacting solution. An acidic solution, for example, will generally work as the reacting solution for most of the O_2 and CO_2 precursor compounds. Manganese dioxide works as a catalyst for the peroxygens.

For the production of oxygen, the present invention involves the use of a group of chemical compounds generally referred to as oxygen precursors, that is, compounds which under certain chemical conditions give off oxygen as an end product. Hydrogen peroxide (H_2O_2) is an example of an oxygen precursor and is suitable for use in the present invention. However, for the present invention, there are other chemical compounds far more suitable and easier to handle than H_2O_2 .

A preferred group of compounds for use in the present invention for the production of oxygen are known as peroxy or peroxygen compounds, and are well known in the bleach and detergent industry and the health care industry. These types of compounds, under suitable conditions, can be employed in the present invention as convenient oxygen producers. Peroxygen compounds usually are precursors of hydrogen peroxide (H_2O_2),

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which in turn, is the precursor compound of oxygen. Peracids (peroxyacids), their precursors, and their salts are all good sources of peroxygen compounds.

A preferred group of compounds for use in the present invention for the production of carbon dioxide are alkali metal and alkaline earth metal salts of carbonates and bicarbonates, e.g., sodium bicarbonate. These types of compounds, under suitable conditions can be employed in the present invention as convenient carbon dioxide producers.

As mentioned above, peroxygen compounds have been in use or considered as oxidizing agents by the bleach and detergent industry and the health care industry. The oxidizing power of these agents comes from their production and release of "active oxygen". This usually means oxygen in its so-called "singlet" state when the oxygen possesses a much higher oxidation potential, which, therefore, produces a much stronger oxidizing or bleaching action than plain O_2 . Plain O_2 gas has only a very small amount of oxidizing power, nevertheless, it often can aid in the peroxy compounds bleaching action by means of the so-called effervescent effect. However, the amount is of no practical value or pertinence to the present invention.

In the present invention, plain gas, e.g., oxygen or carbon dioxide, are the only products of concern. A primary use of the present invention is for aiding or sustaining respiration of living plants or animals. However, the present invention can be employed in any environment where a continuing supply of

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oxygen or carbon dioxide is desired. For example, one area where a need exists for such a system is fish which are being used for bait. Currently, a sufficient supply of oxygen is often unavailable to sustain the fish while they are stored and transported for use as bait. The present invention substantially extends the time over which such bait can be stored and used. Another use is as a temporary source of oxygen for fish in an aquarium or for fish which are being transported from a store to an aquarium. Also, for example, in situations where use of an aeration pump is not possible or practical, the present invention can be employed to provide oxygen. When the present invention is used to provide a source of oxygen for fish, it is important that the pH of the water be maintained at a level acceptable to the fish. One means of doing so is to employ urea peroxide as the oxygen precursor. Another means is to include an additional compound which will maintain a neutral pH, e.g., an acidic calcium salt such as calcium sulfate.

Other possible uses include situations where emergency amounts of oxygen are sometimes required, such as in enclosed areas where the air quality is poor. By employing the present invention, a suitable amount of oxygen can be maintained in the air.

The use of the present invention is not limited to situations where the oxygen is essential to respiration. It can also be employed simply to boost the amount of oxygen already available, such as in rooms where insufficient ventilation is

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available. Another use is to provide additional amounts of oxygen to aid combustion.

The present invention includes the production of inexpensive and simple disposable oxygen-producing or carbon-dioxide-producing units which can be portable, if desired, and/or immersible in liquid, and are also easily replaceable.

Examples of O₂ precursor chemical compounds are:

Sodium perborate anhydrous (theoretical O₂ content 19%)

Sodium perborate monohydrate (theoretical O₂ content 16%)

Sodium perborate tetrahydrate (theoretical O₂ content 10.4%)

Sodium percarbonate
(theoretical O₂ content 15.3%)

Urea peroxide
(theoretical O₂ content 17%)

Peracids such as the various percarboxylic acids:
peroxysuccinic acid and its derivatives.

Diperoxyisophthalic acid

Magnesium monoperoxyphthalate hexahydrate

Potassium monopersulfate

Most of the above compounds produce O₂ when they come in contact with water, with pH and solubility being important

factors in influencing the rate of O_2 production. The use of water will sometimes be sufficient for H_2O_2 precursors such as sodium percarbonate and sodium perborate (anhydrous or monohydrate). When other compounds are employed, such as urea peroxide or the tetrahydrate of sodium perborate, their stability or limited solubility in water may result in an insufficient rate of O_2 production unless a catalytic agent is also employed. Moreover, since the O_2 typically results from decomposition of the initially produced H_2O_2 , catalytic agents which hasten the breakdown of H_2O_2 to O_2 can often be beneficially employed even when compounds are employed which inherently have significant rates of oxygen release. Transition metal salts and complexes, particularly, ethylene diamine tetraacetic acid ("EDTA") chelates are suitable compounds.

The rate of O_2 production can be controlled by varying either the amount or concentration of either of the ingredients. Either of the catalytic agent or the O_2 precursor compounds can be employed in liquid or solid form, provided that a liquid is present to ensure reaction between the ingredients.

The oxygen precursor may be introduced or released in a controlled manner onto the oxygen releasing compound, or alternatively, the oxygen releasing compound may be introduced or released onto the oxygen precursor.

The manner in which catalytic agents increase the rate of oxygen production (release) can be seen in Figs. 1-4. Figs. 1 and 2 demonstrate how the production of oxygen from sodium

perborate can be increased by increasing the concentration of iron EDTA employed as the catalytic agent.

Fig. 3 compares oxygen release from sodium perborate when ferric chloride is used as the catalytic agent versus the use of iron EDTA.

The rate can also be controlled by pelleting or tableting of the oxygen precursor compound with or without a coating and in mixtures of different sizes and/or coatings to moderate the rate of dissolution. Coatings for use in controlling the rate of dissolution are well known to those skilled in the art.

Another way of controlling the rate of O_2 production is having one of the ingredients, e.g., the water (or catalytic solution) or the oxygen precursor compounds released into contact with the other ingredient at a controlled rate. In this manner any specific desired rate can be obtained. Fig. 4 illustrates the controlled release of oxygen from granulated sodium perborate monohydrate and tetrahydrate by the incremental addition of a solution of 0.1M iron EDTA. The arrows indicate the time of addition of the iron EDTA. In each case, 0.1 ml was added, except for the first addition to the monohydrate, where 0.05 ml was added.

Various ways of controlling the feeding rate of a solution are well known to those skilled in the art. For example:

- (1) by using part of the generated gas to effect the

expelling of the liquid,

(2) by the use of a piston operated either by weight or by spraying or gas pressure or by a combination of the preceeding forcing the liquid through a controlled pore size outlet, or

(3) by the capillary action (wicking) of an absorbent material such as cotton.

Fig. 5 illustrates schematically a suitable apparatus for the controlled release of oxygen or carbon dioxide. The opening of tube 10 is covered by microporous hydrophobic plastic cap 12. By using this material for the cap, produced gas can escape while preventing water from entering. At the bottom of tube 10 is tableted oxygen or carbon dioxide precursor, e.g., urea peroxide. To initiate the production of gas, cap 12 is removed and either water or a solution of a catalytic agent is poured into tube 10. Cap 12 is replaced and the production of gas begins, gas bubbles 18 rising through the liquid and then escaping through cap 12 as illustrated by arrows 20, 22 and 24. The apparatus of Fig. 5 is suitable for uses such as supplying oxygen to fish which will be used as bait.

Fig. 6 illustrates schematically another suitable apparatus for the controlled release of gas. External container 30 holds a desired amount of a gas precursor 32. Internal container 34 is filled with liquid 36 which can be water or a catalytic solution depending on the particular gas precursor being employed. Piston 38 assists in the delivery of liquid, and

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can be driven, for example, by its own weight or by the force of a spring (not shown). Liquid 36 is released through outlet 40. Various means can be employed to control the rate at which liquid 36 flows through outlet 40, including the internal diameter of outlet 40, or the use of a porous plug within outlet 40. Porous plug 42 provides a cover for container 30 while still allowing gas 44 to escape. Tube 46 provides for the equalization of pressure in the upper portion of internal container 34 to provide a more even flow rate of liquid 36.

Fig. 7 illustrates schematically yet another suitable apparatus for the controlled release of gas. Container 50 can take various shapes provided that at least one liquid inlet and one gas outlet are provided. In the apparatus of Fig. 7, cotton plugs 52 are located at opposite ends of the apparatus to allow liquid to reach a cotton wick 54 which runs through the center of gas releasing composition 56. A suitable oxygen releasing composition 56 is a mixture of urea peroxide and manganese dioxide. Gas produced by contact of liquid with gas releasing composition 56 exits through microporous plugs 58. Preferred microporous plugs are ceramic and are commercially available.

Fig. 8 is a graph reporting the oxygen release from 15 grams of urea peroxide as a result of water brought into contact by the wicking action of cotton plugs and wicks.

Other suitable designs for the apparatus of Figs. 5, 6, and 7 will be apparent to those skilled in the art.

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In order to further illustrate the present invention and the advantages thereof, the following examples are given, it being understood that these examples are intended only to be illustrative without serving as a limitation on the scope of the present invention.

EXAMPLE 1

To an apparatus, which is weighted to sink in water, as illustrated in Fig. 5, containing 10 grams of urea peroxide is added 25 ml of a 0.02M solution of iron EDTA. The cap is replaced on the tube and the mixture stirred to ensure effective mixing of the ingredients. The apparatus is then placed in a one-gallon bucket of fish bait where oxygen is released at a rate of about 100 ml/hour, for a 9 hour period.

EXAMPLE 2

An apparatus as illustrated in Fig. 7 containing 14 grams of urea peroxide and 0.9 gram of manganese dioxide was placed in a two gallon bait bucket containing two dozen small minnows, i.e., two to three inches long. The minnows were still alive after 10 hours. The by-product urea from the urea peroxide appears to be well tolerated by the fish.

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Although the present invention has been described in terms of various preferred embodiments, one skilled in the art will recognize that various modifications, substitutions, omissions, and changes may be made without departing from the spirit thereof. Accordingly, it is intended that the scope of the present invention be limited only by the scope of the following claims.

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WHAT IS CLAIMED IS:

1. A method of controlled release of a gas comprising the controlled introduction of a specific reacting or catalyzing solution into contact with a precursor compound.
2. The method of claim 1 wherein the gas is oxygen or carbon dioxide.
3. The method of claim 1 wherein the precursor is an oxygen releasing compound.
4. The method of claim 1 wherein the reacting or catalyzing solution is water.
5. The method of claim 1 wherein the reacting or catalyzing solution is acidic.
6. The method of claim 3 wherein the oxygen releasing compound is urea peroxide.
7. A process for supplying oxygen for respiration comprising contacting an oxygen precursor with an oxygen releasing compound.

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8. The process of claim 7 wherein the oxygen releasing compound is water.

9. The process of claim 7 wherein the oxygen releasing compound is a catalytic compound.

10. The process of claim 7 wherein the oxygen precursor is a perborate, a percarbonate, a peroxide, or a peracid.

11. The process of claim 10 wherein the oxygen precursor is sodium perborate, sodium percarbonate, urea peroxide, peroxy succinic acid, diperoxyisophthalic acid, manganese monoperoxyphthalate hexahydrate, or potassium monopersulfate.

12. The process of claim 7 wherein the catalytic compound is in solution and the rate of oxygen release is controlled by the concentration of the catalytic compound.

13. The process of claim 7 wherein the catalytic compound is in solution and the rate of oxygen release is controlled by the rate at which the solution of catalytic compound is supplied to the oxygen precursor.

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14. The process of claim 7 wherein the oxygen precursor is in solid form and the rate of oxygen release is controlled by the dissolution rate of the oxygen precursor.

15. An apparatus for the controlled release of a gas comprising a gas precursor compound, means for controlled introduction of a specific reacting or catalyzing solution, and means for release of gas.

16. The apparatus of claim 15 wherein the means for controlled introduction is means for capillary action.

17. The apparatus of claim 16 wherein the means for capillary action is a cotton wick.

18. The apparatus of claim 15 wherein the means for release of gas is a microporous ceramic plug.

19. The apparatus of claim 15 wherein the gas precursor compound is urea peroxide, the means for controlled introduction of a specific reacting or catalyzing solution is a cotton wick, and the means for release of gas is a microporous ceramic plug.

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20. An apparatus for the controlled release of oxygen comprising an oxygen precursor, an oxygen releasing compound, and means for controlled contacting of the precursor with the oxygen releasing compound.

21. The apparatus of claim 20 wherein the oxygen releasing compound is a catalytic compound.

22. The apparatus of claim 21 wherein the catalytic compound is in solution.

23. The apparatus of claim 20 wherein the solution is released in a controlled manner onto the precursor which is in solid form.

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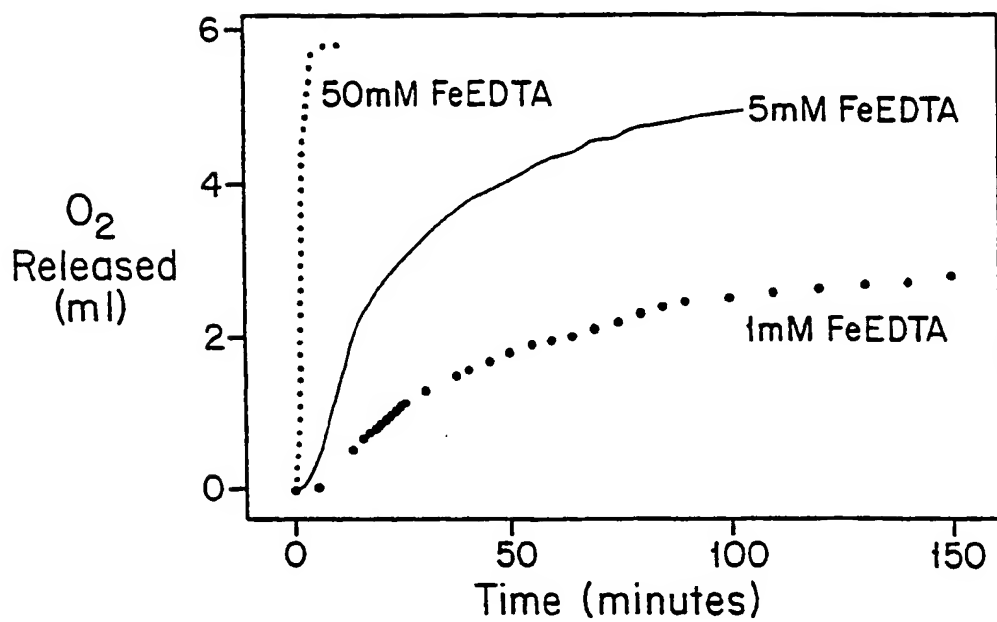
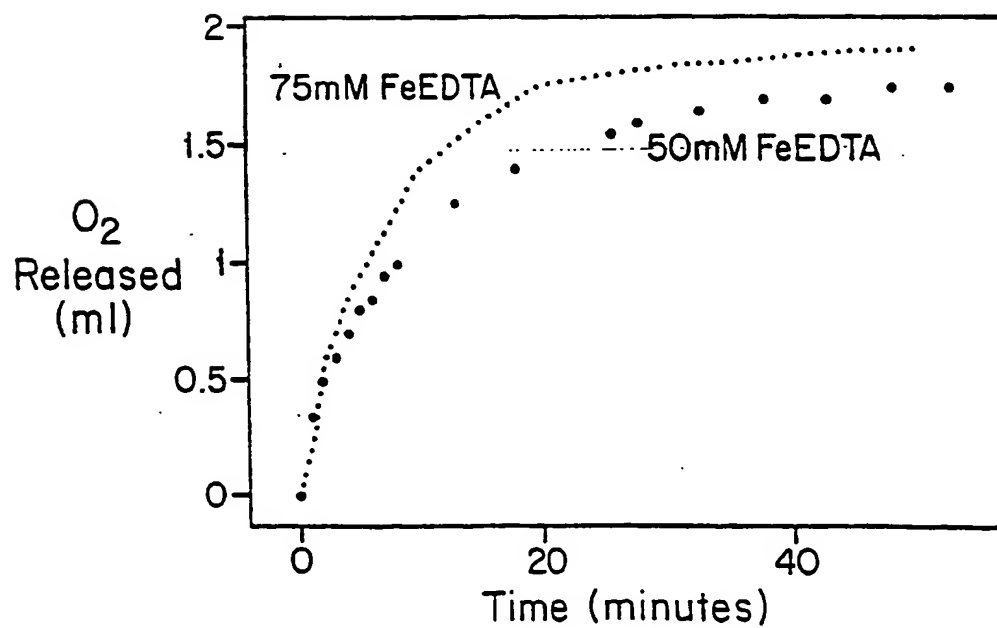
Fig. 1*Fig. 2*

Fig. 3

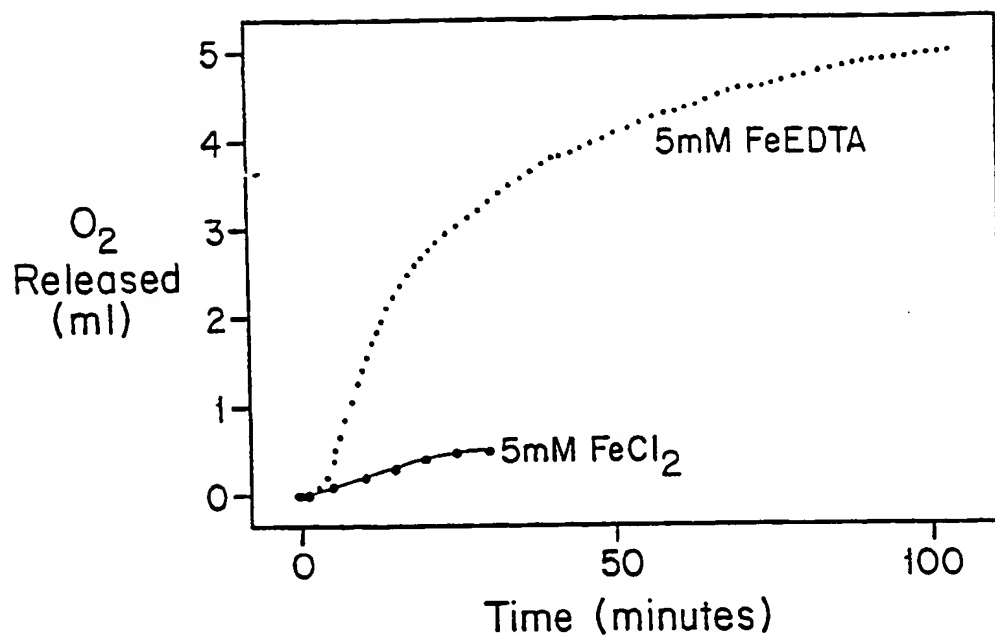
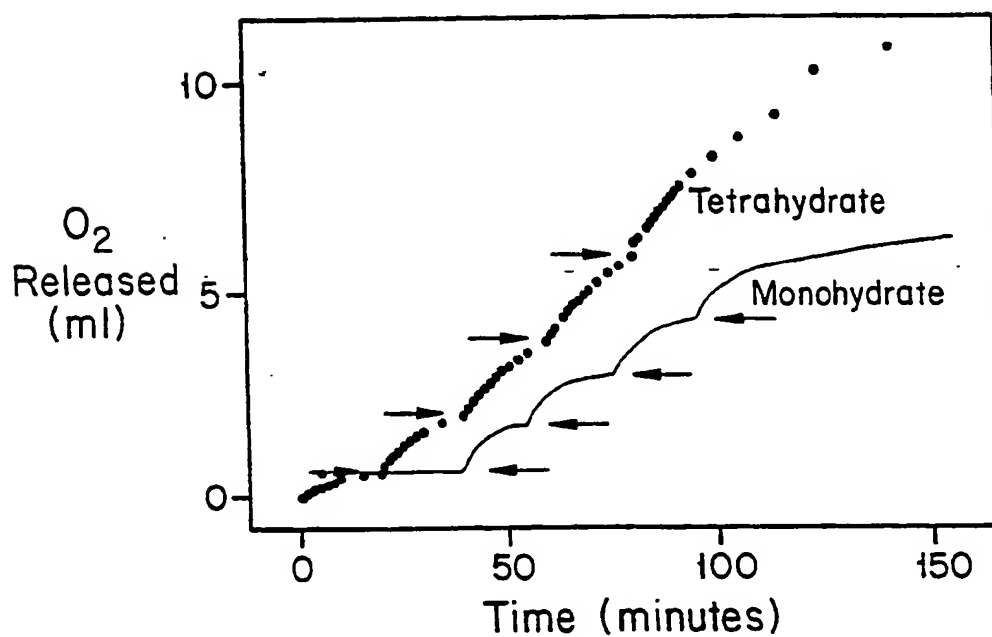
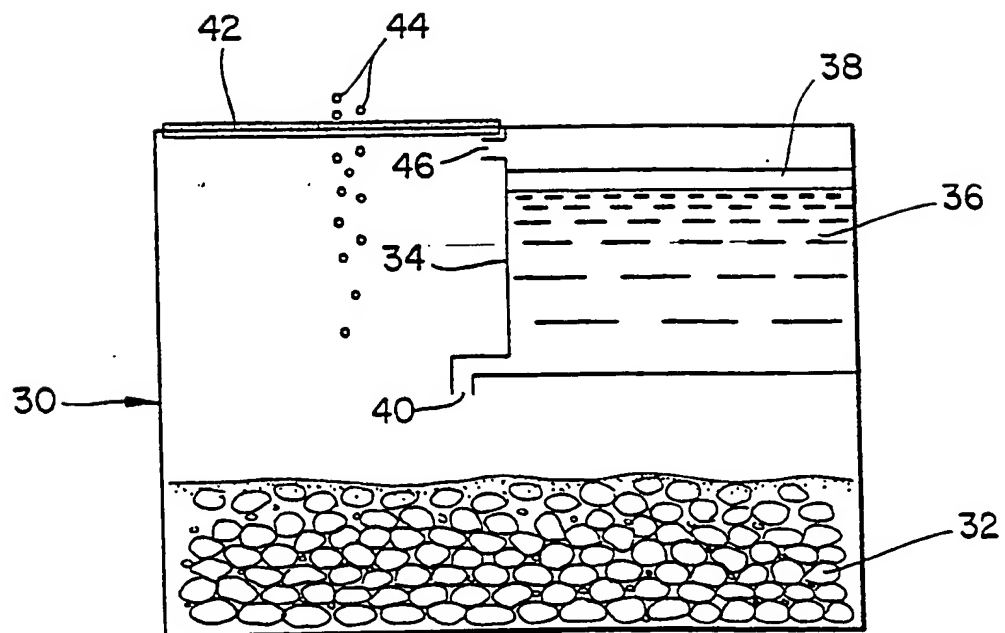
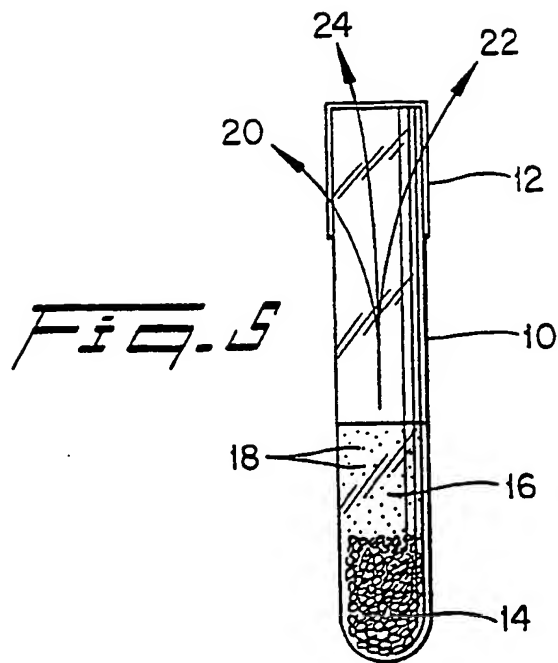


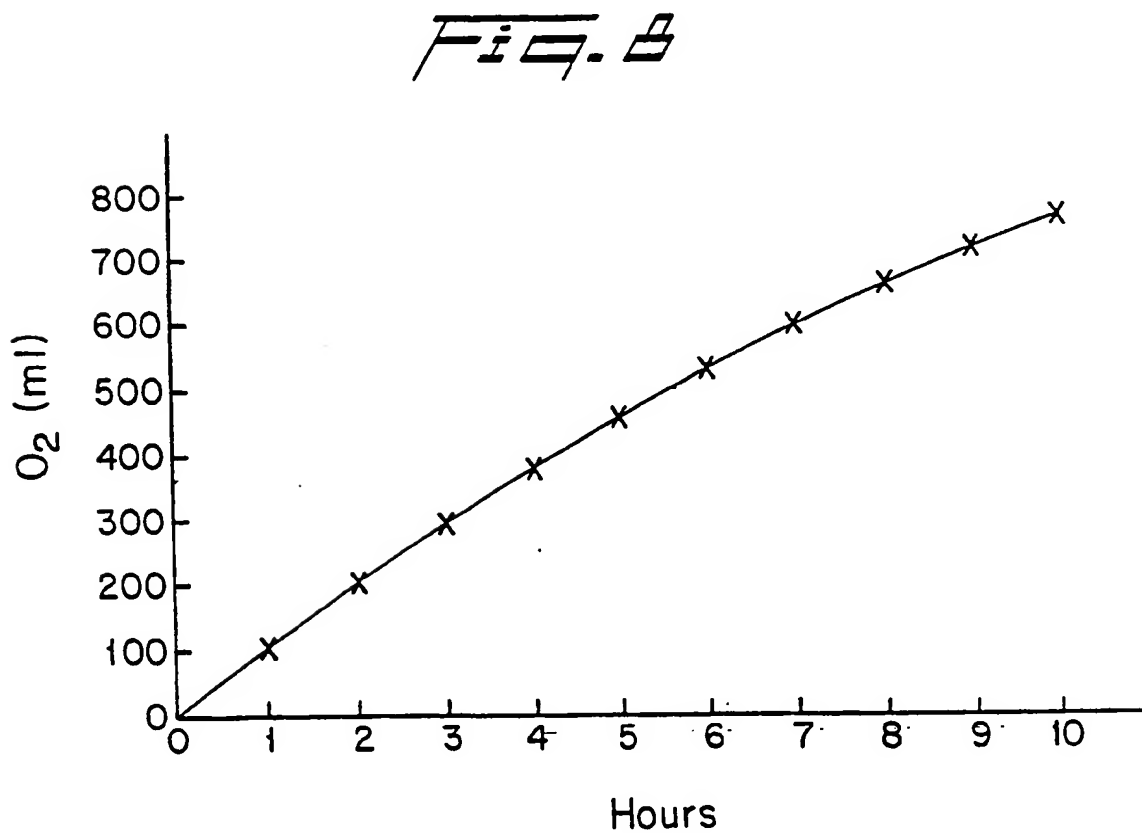
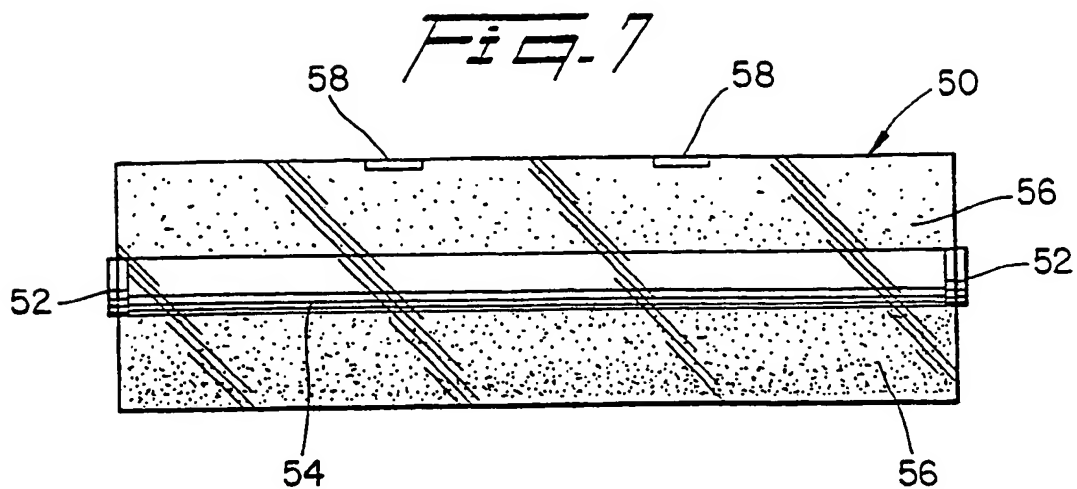
Fig. 4



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*Fig. 6*



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INTERNATIONAL SEARCH REPORT

International Application No. PCT/US90/01523

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶ According to International Patent Classification (IPC) or to both National Classification and IPC <div style="text-align: center; font-family: monospace; font-size: 1.2em;">IPC(5): C01B 13/00; C01B 31/20</div>														
II. FIELDS SEARCHED <div style="text-align: center; font-size: 0.8em;">Minimum Documentation Searched ⁷</div> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 20%; padding: 5px;">Classification System</td> <td style="padding: 5px;">Classification Symbols</td> </tr> <tr> <td style="text-align: center; padding: 10px 20px;">US</td> <td style="padding: 10px 20px;">422/129; 423/437, 579,659</td> </tr> </table> <div style="text-align: center; font-size: 0.8em; margin-top: 5px;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸</div>			Classification System	Classification Symbols	US	422/129; 423/437, 579,659								
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III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹ <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%; padding: 5px;">Category ⁹</th> <th style="width: 60%; padding: 5px;">Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²</th> <th style="width: 30%; padding: 5px;">Relevant to Claim No. ¹³</th> </tr> </thead> <tbody> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">US, A, 4,508,700 (HOSHIKO), 02 April 1985, (See entire document).</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-12</td> </tr> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">US, A, 4,683,130 (VENO ET. AL.), 28 July 1987, (See entire document).</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-12</td> </tr> <tr> <td style="text-align: center; vertical-align: top; padding: 5px;">X</td> <td style="padding: 5px;">US, A, 4,620,970 (SUZUMORI ET. AL.), 04 November 1986, (See entire document).</td> <td style="text-align: center; vertical-align: top; padding: 5px;">1-12</td> </tr> </tbody> </table>			Category ⁹	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³	X	US, A, 4,508,700 (HOSHIKO), 02 April 1985, (See entire document).	1-12	X	US, A, 4,683,130 (VENO ET. AL.), 28 July 1987, (See entire document).	1-12	X	US, A, 4,620,970 (SUZUMORI ET. AL.), 04 November 1986, (See entire document).	1-12
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<div style="display: flex; justify-content: space-between; font-size: 0.8em;"> <div style="width: 45%;"> <p>[*] Special categories of cited documents: ¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p> </div> </div>														
IV. CERTIFICATION <table style="width: 100%;"> <tr> <td style="width: 50%; vertical-align: top;"> Date of the Actual Completion of the International Search <div style="font-family: monospace; font-size: 1.2em;">07 MAY 1990</div> </td> <td style="width: 50%; vertical-align: top;"> Date of Mailing of this International Search Report <div style="font-family: monospace; font-size: 1.2em;">18 JUL 1990</div> </td> </tr> <tr> <td style="vertical-align: top;"> International Searching Authority <div style="font-family: monospace; font-size: 1.2em;">ISA / US</div> </td> <td style="vertical-align: top;"> Signature of Authorized Officer <div style="text-align: center; font-family: monospace; font-weight: bold;">WAYNE A. LANGEL</div> </td> </tr> </table>			Date of the Actual Completion of the International Search <div style="font-family: monospace; font-size: 1.2em;">07 MAY 1990</div>	Date of Mailing of this International Search Report <div style="font-family: monospace; font-size: 1.2em;">18 JUL 1990</div>	International Searching Authority <div style="font-family: monospace; font-size: 1.2em;">ISA / US</div>	Signature of Authorized Officer <div style="text-align: center; font-family: monospace; font-weight: bold;">WAYNE A. LANGEL</div>								
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